

Windings of the 2D free Rouse chain

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Abstract

We study long time dynamical properties of a chain of harmonically bound Brownian particles. This chain is allowed to wander everywhere in the plane. We show that the scaling variables for the occupation times T_j , areas A_j and winding angles θ_j ($j = 1, \dots, n$ labels the particles) take the same general form as in the usual Brownian motion. We also compute the asymptotic joint laws $P(\{T_j\})$, $P(\{A_j\})$, $P(\{\theta_j\})$ and discuss the correlations occurring in those distributions.

1 Introduction

In order to study the dynamics of dilute polymer solutions, P.E. Rouse proposed in 1953 his famous model of harmonically bound Brownian particles (Rouse chain) [1]. Since that time, this model has become very popular in the field of polymer science. It appears that, despite its drawbacks and limitations (in particular, absence of excluded volume and hydrodynamic interactions), it is conceptually important and useful to study the dynamics of polymers in melts [2,3]. In this paper, we will consider the free planar motion of such a chain of n particles (monomers) and especially address its long time ($t \rightarrow \infty$) properties from the Brownian motion viewpoint.

A configuration of this chain being represented by a complex n -vector z (the components z_i , $i = 1, \dots, n$ are the complex coordinates of the particles), we will study closed

trajectories of length t , i.e. $z(t) = z(0)$ or open ones ($z(0)$ fixed, $z(t)$ left unspecified, i.e. integrated over).

More precisely, if we consider some given bounded domain S of area \mathcal{S} and define the occupation time T_j as the time spent inside S by the j^{th} particle, our goal is to compute the joint probability distribution $P(T_1, T_2, \dots, T_n) (\equiv P(\{T_j\}))$. Similarly, A_j and θ_j being respectively the area enclosed by the trajectory of the j^{th} particle and its winding angle around O , we will be interested in the joint laws $P(\{A_j\})$ and $P(\{\theta_j\})$.

On general grounds, we expect that the various properties of the chain will be strongly influenced by the free Brownian motion of the center of mass (c.o.m.) of the chain. But will they strictly satisfy the same laws ? With the same scaling variables ? And what about the correlations among the different variables ? Before answering those questions, we first recall some standard results concerning a planar Brownian particle with a diffusion constant D [4-8].

Results i) and ii) concern open trajectories when $t \rightarrow \infty$ while iii) concerns closed trajectories and is valid for all times t :

- i) Kallianpur-Robbins' law [4] for the probability distribution of the occupation time T of a bounded domain of area \mathcal{S} :

$$P\left(T' = \frac{4\pi D T}{\mathcal{S} \ln t}\right) = \theta(T') e^{-T'} \quad (1)$$

- ii) Spitzer's law [5] for the angle θ wound around a given point:

$$P\left(\theta' = \frac{2\theta}{\ln t}\right) = \frac{1}{\pi} \frac{1}{1 + (\theta')^2} \quad (2)$$

with the characteristic function:

$$\langle e^{i\lambda\theta'} \rangle = e^{-|\lambda|} \quad (3)$$

- iii) Lévy's law [6] for the area A enclosed by the closed trajectory of the particle:

$$P\left(A' = \frac{A}{2 D t}\right) = \frac{\pi}{2} \frac{1}{\cosh^2(\pi A')} \quad (4)$$

$$\langle e^{iBA'} \rangle = \frac{\left(\frac{B}{2}\right)}{\sinh\left(\frac{B}{2}\right)} \quad (5)$$

The distributions i) and iii) have moments of all orders in contrast with ii) that has none.

Those laws were discovered more than 40 years ago and since that time, many refinements have been made. For instance, in [7], the authors found the asymptotic ($t \rightarrow \infty$)

joint law of the small (θ_-) and big (θ_+) windings. θ_- (resp. θ_+) are the angles wound around O and only counted when r is smaller (resp. greater) than some fixed r_0 (r is the distance separating the particle from O). With the rescaled angles $\theta'_\pm = \frac{2\theta_\pm}{\ln t}$, the characteristic function writes [7]:

$$\left\langle e^{i(\lambda_- \theta'_- + \lambda_+ \theta'_+)} \right\rangle = \frac{1}{\cosh(\lambda_+) + \frac{|\lambda_-|}{\lambda_+} \sinh(\lambda_+)} \quad (6)$$

($\lambda_+ = \lambda_- = \lambda$ gives back Spitzer's law (2)).

Remark that (2) and (6) don't depend on the diffusion constant. This is quite different from the Brownian motion on a bounded domain surrounding O. In that case, we have [9]:

$$\left\langle e^{i\lambda\theta} \right\rangle = e^{-cD|\lambda|t} \quad (7)$$

where c is a constant depending on the geometry and the boundary conditions. Here, D , appears as a multiplier of $|\lambda|$. We will use this remark at the end of the paper.

2 The free Rouse chain

Starting our study, we consider the following set of coupled Langevin equations:

$$\begin{aligned} \dot{z}_1 &= k(z_2 - z_1) + \eta_1 \\ \dot{z}_l &= k(z_{l+1} + z_{l-1} - 2z_l) + \eta_l, \quad 2 \leq l \leq n-1 \\ \dot{z}_n &= k(z_{n-1} - z_n) + \eta_n \end{aligned} \quad (8)$$

where k is the spring constant and η_m ($\equiv \eta_{mx} + i\eta_{my}$) a gaussian white noise:

$$\begin{aligned} \langle \eta_m(t) \rangle &= 0 \\ \langle \eta_m(t) \eta_{m'}(t') \rangle &= 2\delta_{mm'} \delta(t - t') \end{aligned} \quad (9)$$

(This noise would correspond to a $D = 1/2$ diffusion constant if particles were free).

For the chain c.o.m., we get $\dot{z}_G = \frac{1}{n}(\sum_{i=1}^n \eta_i) \equiv \eta_G$ with $\langle \eta_G(t) \eta_G(t') \rangle = \frac{2}{n} \delta(t - t')$. The c.o.m. motion is free with $D = 1/(2n)$.

Introducing the complex n -vector η , eq.(8) can be written in a matrix form:

$$\dot{z} = -k \mathbf{M} z + \eta \quad (10)$$

where \mathbf{M} is the tridiagonal ($n \times n$) matrix:

$$\mathbf{M} = \begin{pmatrix} 1 & -1 & 0 & \cdots & 0 \\ -1 & 2 & -1 & \cdots & 0 \\ 0 & -1 & 2 & \cdots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \cdots & 1 \end{pmatrix}$$

with eigenvalues:

$$\omega_j = 2 \left(1 - \cos \frac{\pi(j-1)}{n} \right), \quad 1 \leq j \leq n \quad (11)$$

$$(\omega_1 = 0; \det' \mathbf{M} \equiv \prod_{j=2}^n \omega_j = n)$$

With the matrix $\omega = \text{diag}(\omega_i)$, we can write:

$$\omega = \mathbf{R}^{-1} \mathbf{M} \mathbf{R} \quad (12)$$

$$z = \mathbf{R} Z \quad (13)$$

where \mathbf{R} is an orthogonal matrix and the components of Z are the normal coordinates, that we will widely use in the sequel. From $\mathbf{R}_{j1} = \frac{1}{\sqrt{n}}$, $j = 1, \dots, n$, we deduce that $Z_1 (= \sum_{i=1}^n z_i / \sqrt{n})$ is essentially the c.o.m. coordinate. Remark also that $\sum_{i=2}^n \omega_i |Z_i|^2 = \sum_{i=2}^n |z_i - z_{i-1}|^2 = {}^t \bar{z} \mathbf{M} z$.

Let us call $\mathcal{P}(z, z^{(0)}, t)$ the probability for the chain to go from configuration $z^{(0)}$ at $t = 0$ to z at time t . \mathcal{P} satisfies a Fokker-Planck equation [10]:

$$\partial_t \mathcal{P} = \left({}^t \partial_z k \mathbf{M} z + {}^t \partial_{\bar{z}} k \mathbf{M} \bar{z} + 2 {}^t \partial_{\bar{z}} \partial_z \right) \mathcal{P} \quad (14)$$

where ∂_z (resp. $\partial_{\bar{z}}$) is a n -vector of components ∂_{z_i} (resp. $\partial_{\bar{z}_i}$) and ${}^t \partial_z$ (resp. ${}^t \partial_{\bar{z}}$) is the transpose of ∂_z (resp. $\partial_{\bar{z}}$). The solution can be written in terms of a path integral ($\mathcal{D}z \mathcal{D}\bar{z} = \prod_{i=1}^n \mathcal{D}z_i \mathcal{D}\bar{z}_i$):

$$\begin{aligned} \mathcal{P}(z, z^{(0)}, t) &= \det \left(e^{tk\mathbf{M}} \right) \int_{z^{(0)}}^z \mathcal{D}z \mathcal{D}\bar{z} \exp \left(-\frac{1}{2} \int_0^t {}^t (\dot{\bar{z}} + k\mathbf{M}\bar{z})(\dot{z} + k\mathbf{M}z) d\tau \right) \\ &\equiv F(z, z^{(0)}) \cdot G_0(z, z^{(0)}, t) \end{aligned} \quad (15)$$

with

$$\begin{aligned} F(z, z^{(0)}) &= e^{-\frac{k}{2} ({}^t \bar{z} \mathbf{M} z - {}^t \bar{z}^{(0)} \mathbf{M} z^{(0)})} = \\ &= e^{-\frac{k}{2} \sum_{i=2}^n (|z_i - z_{i-1}|^2 - |z_i^{(0)} - z_{i-1}^{(0)}|^2)} = e^{-\frac{k}{2} \sum_{i=2}^n \omega_i (|Z_i|^2 - |Z_i^{(0)}|^2)} \\ G_0(z, z^{(0)}, t) &= \int_{z^{(0)}}^z \mathcal{D}z \mathcal{D}\bar{z} \exp \left(-\frac{1}{2} \int_0^t \left({}^t \dot{\bar{z}} \dot{z} + k^2 {}^t \bar{z} \mathbf{M}^2 z - 2k \text{Tr} \mathbf{M} \right) d\tau \right) = \\ &= \left\langle z \left| e^{-tH_0} \right| z^{(0)} \right\rangle \end{aligned} \quad (16)$$

$$H_0 = -2 {}^t \partial_{\bar{z}} \partial_z + \frac{1}{2} k^2 {}^t \bar{z} \mathbf{M}^2 z - k \text{Tr} \mathbf{M} \quad (17)$$

In fact, \mathcal{P} , eq.(15), can be easily deduced from the gaussian distribution of η (use (10); $\det(e^{tk\mathbf{M}})$ is the functional Jacobian for the change of variable $\eta \rightarrow z$ [11]).

$G_0(z, z^{(0)}, t)$ is most conveniently written in terms of the normal coordinates Z_i and $Z_i^{(0)}$, clearly exhibiting the free motion of the c.o.m. [12]:

$$G_0(z, z^{(0)}, t) = \frac{1}{2\pi t} e^{-\frac{1}{2t}|Z_1 - Z_1^{(0)}|^2} \times \\ \times \prod_{i=2}^n \left(\frac{s_i e^{k\omega_i t}}{2\pi} \exp \left\{ -\frac{1}{2} \left(\bar{Z}_i c_i Z_i + \bar{Z}_i^{(0)} c_i Z_i^{(0)} - \bar{Z}_i^{(0)} s_i Z_i - \bar{Z}_i s_i Z_i^{(0)} \right) \right\} \right) \quad (18) \\ s_i = \frac{k\omega_i}{\sinh(k\omega_i t)} ; c_i = k\omega_i \coth(k\omega_i t)$$

When $kt \gg 1$, we get, for G_0 , the limiting expression:

$$G_0^\infty(z, z^{(0)}, t) = \frac{1}{2\pi t} e^{-\frac{1}{2t}|Z_1 - Z_1^{(0)}|^2} \prod_{i=2}^n \left(\frac{k\omega_i}{\pi} e^{-\frac{k\omega_i}{2}(|Z_i|^2 + |Z_i^{(0)}|^2)} \right) \quad (19) \\ \equiv \mathcal{G}_0(z, z^{(0)}, t) \cdot g_0(z, z^{(0)})$$

where \mathcal{G}_0 is the c.o.m. propagator and g_0 can be simply written in terms of the z_i :

$$g_0(z, z^{(0)}) = n \left(\frac{k}{\pi} \right)^{n-1} e^{-\frac{k}{2} \sum_{i=2}^n (|z_i - z_{i-1}|^2 + |z_i^{(0)} - z_{i-1}^{(0)}|^2)} \quad (20)$$

Furthermore, as can be easily checked, \mathcal{P} is properly normalized:

$$\int dz d\bar{z} \mathcal{P}(z, z^{(0)}, t) = 1 \text{ (} G_0 \text{ given by (18) or (19)).}$$

Now, we turn to the computation of the joint law $P(\{T_j\})$.

3 Occupation times distribution

Recall that T_j is the time spent by particle j inside a bounded domain S of area \mathcal{S} . We consider trajectories starting at $t = 0$ from some given configuration $z^{(0)}$ and reaching at time t the final configuration z . Leaving z unspecified, we have, with positive p_i 's:

$$\langle e^{-\sum_{i=1}^n p_i T_i} \rangle = \det(e^{tk\mathbf{M}}) \times \\ \times \int dz d\bar{z} \int_{z^{(0)}}^z \mathcal{D}z \mathcal{D}\bar{z} \exp \left(- \int_0^t \left(\frac{1}{2} \dot{\bar{z}} (\dot{z} + k\mathbf{M}\bar{z}) (\dot{z} + k\mathbf{M}z) + V_P(z) \right) d\tau \right) \quad (21)$$

$$= \int dz d\bar{z} F(z, z^{(0)}) G_P(z, z^{(0)}, t) \quad (22)$$

with

$$G_P(z, z^{(0)}, t) = \langle z \mid e^{-t(H_0 + V_P)} \mid z^{(0)} \rangle \quad (23)$$

$$V_P(z) = \sum_{i=1}^n p_i \mathbf{1}_S(z_i) \quad (24)$$

$\mathbf{1}_S(z_i)$ is the indicatrix function of the domain S . Symbolically, we write:

$$G_P = \sum_{m=0}^{\infty} (-1)^m G_0 (V_P G_0)^m \quad (25)$$

with

$$\begin{aligned} G_0 (V_P G_0)^m &= \int_0^t dt_m \int_0^{t_m} dt_{m-1} \dots \int_0^{t_2} dt_1 \int \left(\prod_{j=1}^m d\bar{z}^{(j)} dz^{(j)} \right) G_0(z, z^{(m)}, t - t_m) \times \\ &\times V_P(z^{(m)}) G_0(z^{(m)}, z^{(m-1)}, t_m - t_{m-1}) V_P(z^{(m-1)}) \dots V_P(z^{(1)}) G_0(z^{(1)}, z^{(0)}, t_1) \end{aligned} \quad (26)$$

$(z^{(j)})$ is the chain configuration at time t_j ; $d\bar{z}^{(j)} dz^{(j)} = \prod_{i=1}^n d\bar{z}_i^{(j)} dz_i^{(j)}$.

Let us compute the contribution $N_m(t)$ of this generic term to (22). Integrating over z , we have:

$$\begin{aligned} N_m(t) &= (-1)^m \int_0^t dt_m \int_0^{t_m} dt_{m-1} \dots \int_0^{t_2} dt_1 \int \left(\prod_{j=1}^m d\bar{z}^{(j)} dz^{(j)} \right) F(z^{(m)}, z^{(0)}) \times \\ &\times V_P(z^{(m)}) G_0(z^{(m)}, z^{(m-1)}, t_m - t_{m-1}) \dots V_P(z^{(1)}) G_0(z^{(1)}, z^{(0)}, t_1) \end{aligned} \quad (27)$$

$$\equiv (-1)^m \int_0^t dt_m \int \left(\prod_{j=1}^m d\bar{z}^{(j)} dz^{(j)} V_P(z^{(j)}) \right) F(z^{(m)}, z^{(0)}) \Phi(t_m, \{z^{(l)}\}) \quad (28)$$

Φ is a time convolution product of the free propagators G_0 . Disregarding for the moment the spatial integrations, the above expression is well-suited, in the limit $t \rightarrow \infty$, for applying Tauberian theorems [13]. Introducing the Laplace Transform $\hat{\Phi}$:

$$\hat{\Phi}(u, \{z^{(l)}\}) \equiv \int_0^\infty e^{-ut'} \Phi(t', \{z^{(l)}\}) dt' = \prod_{k=1}^m \hat{G}_0(z^{(k)}, z^{(k-1)}, u) \quad (29)$$

we notice that, when $u \rightarrow 0^+$:

$$\hat{G}_0(z^{(k)}, z^{(k-1)}, u) \equiv \int_0^\infty e^{-ut'} G_0(z^{(k)}, z^{(k-1)}, t') dt' \sim \int_a^\infty e^{-ut'} G_0(z^{(k)}, z^{(k-1)}, t') dt' \quad (30)$$

for some large a . So, we can use the asymptotic form G_0^∞ in the computation of \widehat{G}_0 and get:

$$\widehat{G}_0(z^{(k)}, z^{(k-1)}, u) \sim_{u \rightarrow 0^+} \ln\left(\frac{1}{u}\right) \frac{1}{2\pi} g_0(z^{(k)}, z^{(k-1)}) \quad (31)$$

A weak Tauberian theorem [13] gives for the time integration in (28):

$$\int_0^t dt_m \Phi(t_m, \{z^{(l)}\}) \sim_{t \rightarrow \infty} \left(\frac{\ln t}{2\pi}\right)^m \prod_{k=1}^m g_0(z^{(k)}, z^{(k-1)}) \quad (32)$$

Finally, the result for $N_m(t)$ is:

$$N_m(t) \sim_{t \rightarrow \infty} (-1)^m \left(\frac{n L_P \ln t}{2\pi}\right)^m \quad (33)$$

$$L_P = \int \left(\prod_{i=1}^n dz_i d\bar{z}_i\right) V_P(z) \left(\frac{k}{\pi}\right)^{n-1} e^{-k \sum_{i=2}^n |z_i - z_{i-1}|^2} \quad (34)$$

L_P is computed with V_P , eq.(24): $L_P = (\sum_{i=1}^n p_i) \mathcal{S}$

Rescaling the occupation times T_i :

$$T'_i = \frac{2\pi T_i}{n \mathcal{S} \ln t} \quad (35)$$

we get:

$$\langle e^{-\sum_{i=1}^n p_i T'_i} \rangle = \sum_{m=0}^{\infty} (-1)^m \left(\sum_{i=1}^n p_i\right)^m = \frac{1}{1 + (\sum_{i=1}^n p_i)} \quad (36)$$

This relationship is actually valid, by analytic continuation, for all the positive p_i 's (and not only when $\sum p_i < 1$). This is because the distribution $P(\{T_j\})$ has moments of all orders and consequently $\langle e^{-\sum_{i=1}^n p_i T_i} \rangle$ is holomorphic when $\text{Re}(p_i) \geq 0$.

(36) leads to the probability distribution:

$$P(\{T'_i\}) = \theta(T'_1) e^{-T'_1} \prod_{i=2}^n \delta(T'_i - T'_{i-1}) \quad (37)$$

($n = 1$ gives back the Kallianpur-Robbins' law).

So, in the large time limit, the T_i 's are strongly correlated leading to identical (T'_i) 's. Moreover, we remark that T_i scales like n and, also, that the law for the c.o.m. would be the same as for one monomer (compare (35) to - (1) with $D = 1/(2n)$): the c.o.m. free motion dominates this process.

We also got similar exponential distributions for the rescaled variables T' in the following cases:

- i) T is the time spent when the *whole* chain is inside S . L_P , eq.(34), is now computed with $V_P(z) = p(\prod_{i=1}^n \mathbf{1}_S(z_i))$. Introducing

$$w(\mathcal{S}) = \int \left(\prod_{i=1}^n dz_i d\bar{z}_i \mathbf{1}_S(z_i) \right) e^{-k \sum_{i=2}^n |z_i - z_{i-1}|^2} \quad (38)$$

the rescaled variable writes:

$$T' = \left(\frac{\pi}{k} \right)^{n-1} \frac{2 \pi T}{n w(\mathcal{S}) \ln t} \quad (39)$$

In contrast with (35), k is now present in the asymptotic law. For instance, if S is a small disk of radius r_0 ($kr_0^2 \ll 1$), then $w(\mathcal{S}) \sim \mathcal{S}^n$ and T scales like k^{n-1} : when k grows, the chain collapses and it is easier to confine it inside a given domain.

- ii) T is the time spent by the chain when *at least* one of its particles is inside S . T' is similar to (39) except that $w(\mathcal{S})$ must be changed: L_P is now computed with $V_P(z) = p(1 - \prod_{i=1}^n (1 - \mathbf{1}_S(z_i)))$.

To conclude this section, let us draw two lessons:

- i) The scaling variables take the same general form as for the free Brownian particle. In the sequel, we will show that it is still true for the other quantities we study.
- ii) For the computation of the perturbation theory, when $t \rightarrow \infty$, we can systematically use the asymptotic form G_0^∞ of the unperturbed propagator. Obviously, for this consideration to hold, we must be sure that the perturbation series is well behaved. In those conditions, we will make a wide use of this remark.

4 Areas distribution

Now, we compute the areas distribution $P(\{A_j\})$ for closed trajectories of length t starting and ending at some fixed $z^{(0)}$. To do so, we insert the constraint:

$$\prod_{j=1}^n \delta \left(A_j - \frac{1}{4i} \int_0^t (z_j \dot{\bar{z}}_j - \bar{z}_j \dot{z}_j) d\tau \right) \quad (40)$$

in the measure (15) and use the relationship $\delta(x) = \frac{1}{2\pi} \int e^{iBx} dB$. It is easy to show that this manipulation amounts to add n different magnetic fields B_j to the initial system. Those fields are uniform, orthogonal to the motion plane and such that particle j is submitted to B_j .

With the $(n \times n)$ diagonal matrix \mathbf{B} ($\mathbf{B}_{ij} = B_i \delta_{ij}$), we get

$$P(\{A_i\}) = \int \left(\prod_{j=1}^n \frac{dB_j}{2\pi} e^{iB_j A_j} \right) \left(\frac{G_{\mathbf{B}}(z^{(0)}, z^{(0)}, t)}{G_0(z^{(0)}, z^{(0)}, t)} \right) \quad (41)$$

$$\text{with } G_{\mathbf{B}}(z^{(0)}, z^{(0)}, t) = \langle z^{(0)} | e^{-tH_{\mathbf{B}}} | z^{(0)} \rangle \quad (42)$$

$$H_{\mathbf{B}} = H_0 + V_{\mathbf{B}} \quad (43)$$

$$V_{\mathbf{B}}(z) = \frac{1}{2} \left(-{}^t z \mathbf{B} \partial_z + {}^t \bar{z} \mathbf{B} \partial_{\bar{z}} \right) + \frac{1}{8} {}^t \bar{z} \mathbf{B}^2 z \quad (44)$$

$$= \frac{1}{2} \left(-{}^t Z \mathbf{B}' \partial_Z + {}^t \bar{Z} \mathbf{B}' \partial_{\bar{Z}} \right) + \frac{1}{8} {}^t \bar{Z} \mathbf{B}'' Z \quad (45)$$

($\mathbf{B}' = \mathbf{R}^{-1} \mathbf{B} \mathbf{R}$, $\mathbf{B}'' = \mathbf{R}^{-1} \mathbf{B}^2 \mathbf{R}$). In principle, $P(\{A_j\})$ depends on $z^{(0)}$ but we will show that, actually, this is not the case when $t \rightarrow \infty$ (remark that, for all t , $P(\{A_j\})$ doesn't depend on the c.o.m. of $z^{(0)}$: this is due to translation invariance and this is the reason why we consider the propagator and not the partition function that would diverge like the area of the plane, leading to serious problems in the perturbation theory).

Now, let us sketch the perturbative computation of $G_{\mathbf{B}}$. Following our previous remarks, we will use G_0^∞ for the unperturbed propagator. The generic term writes:

$$\begin{aligned} & (-1)^m \int_0^t dt_m \int_0^{t_m} dt_{m-1} \dots \int_0^{t_2} dt_1 \int \left(\prod_{j=1}^m d\bar{z}^{(j)} dz^{(j)} \right) \dots \\ & \dots G_0^\infty(z^{(j+1)}, z^{(j)}, t_{j+1} - t_j) V_{\mathbf{B}}(z^{(j)}) G_0^\infty(z^{(j)}, z^{(j-1)}, t_j - t_{j-1}) \dots \end{aligned} \quad (46)$$

With normal coordinates, $V_{\mathbf{B}}(z^{(j)})$ takes the form:

$$V_{\mathbf{B}}(z^{(j)}) = \sum_{i,l=1}^n \left(\frac{1}{2} \left(-Z_i^{(j)} \mathbf{B}'_{il} \partial_{Z_l^{(j)}} + \bar{Z}_i^{(j)} \mathbf{B}'_{il} \partial_{\bar{Z}_l^{(j)}} \right) + \frac{1}{8} \bar{Z}_i^{(j)} \mathbf{B}''_{il} Z_l^{(j)} \right) \quad (47)$$

We now proceed by inspection:

- i) For the terms $Z_i^{(j)} \mathbf{B}'_{il} \partial_{Z_l^{(j)}}$, only will survive, after integration, those contributions with $i = j$. The same holds for $\bar{Z}_i^{(j)} \mathbf{B}'_{il} \partial_{\bar{Z}_l^{(j)}}$. Moreover, the diagonal contributions exactly cancel except when $i = j = 1$.
- ii) We reach the same conclusion for the terms $\bar{Z}_i^{(j)} \mathbf{B}''_{il} Z_l^{(j)}$ (non diagonal terms vanish after integration. Diagonal contributions, $i = j > 1$, are subleading – compared to $i = j = 1$ – when $kt \gg 1$: we recover the fact that the process is dominated by the c.o.m. motion).

We are finally left with an effective perturbation $V_{\mathbf{B}}^{\text{eff}}$:

$$V_{\mathbf{B}}^{\text{eff}} = \frac{1}{2} \left(-Z_1 \mathbf{B}'_{11} \partial_{Z_1} + \bar{Z}_1 \mathbf{B}'_{11} \partial_{\bar{Z}_1} \right) + \frac{1}{8} \mathbf{B}''_{11} |Z_1|^2 \quad (48)$$

Only the first mode is affected by the magnetic fields and we can disregard the other modes that will cancel when taking the ratio $G_{\mathbf{B}}/G_0$.

Remark that

$$\mathbf{B}'_{11} = \frac{1}{n} \left(\sum_{i=1}^n B_i \right) \text{ and } \mathbf{B}''_{11} = \frac{1}{n} \left(\sum_{i=1}^n B_i^2 \right) \quad (49)$$

The effective hamiltonian for the remaining mode writes:

$$H_{\mathbf{B}}^{\text{eff}} = -2\partial_{Z_1}\partial_{\bar{Z}_1} + \frac{1}{2} \left(-Z_1\mathbf{B}'_{11}\partial_{Z_1} + \bar{Z}_1\mathbf{B}'_{11}\partial_{\bar{Z}_1} + \frac{1}{4}(\mathbf{B}'_{11})^2|Z_1|^2 \right) + \frac{1}{8} \left(\mathbf{B}''_{11} - (\mathbf{B}'_{11})^2 \right) |Z_1|^2 \quad (50)$$

It describes the behavior of a charged particle submitted to an uniform magnetic field \mathbf{B}'_{11} and an harmonic oscillator of frequency $\omega = \frac{1}{2}\sqrt{\mathbf{B}''_{11} - (\mathbf{B}'_{11})^2}$. Using known results about this problem [14], we immediately get:

$$\begin{aligned} \frac{G_{\mathbf{B}}(z^{(0)}, z^{(0)}, t)}{G_0(z^{(0)}, z^{(0)}, t)} &= \frac{t\sqrt{\mathbf{B}''_{11}}}{2 \sinh\left(\frac{t}{2}\sqrt{\mathbf{B}''_{11}}\right)} \times \\ &\times \exp\left(-\frac{\sqrt{\mathbf{B}''_{11}} \left(\cosh\left(\frac{t}{2}\sqrt{\mathbf{B}''_{11}}\right) - \cosh\left(\frac{t}{2}\mathbf{B}'_{11}\right) \right) |Z_1^{(0)}|^2}{2 \sinh\left(\frac{t}{2}\sqrt{\mathbf{B}''_{11}}\right)} \right) \end{aligned} \quad (51)$$

However, for our computation to be consistent, we must consider this expression in the large time limit. This is readily done in rescaling the areas $A'_i = A_i/t$ and doing $t \rightarrow \infty$. The final expression for the characteristic function of $P(\{A'_i\})$ is quite simple:

$$\left\langle e^{i \sum_{j=1}^n B_j A'_j} \right\rangle = \frac{\sqrt{\mathbf{B}''_{11}}}{2 \sinh\left(\frac{\sqrt{\mathbf{B}''_{11}}}{2}\right)} \quad (52)$$

($\mathbf{B}''_{11} = \frac{1}{n} \sum_{i=1}^n B_i^2$; when $n = 1$, (52) gives back Lévy's result, eq.(5)).

Owing to the form of \mathbf{B}''_{11} , $P(\{A'_i\})$ will only be a function of the variable $\sqrt{\sum_{i=1}^n (A'_i)^2} (\equiv A')$, showing clearly that the (A'_i) 's are correlated. Its determination is reduced to the computation of the following integral [13]:

$$P(\{A'_i\}) \equiv P(A') = \left(\frac{2n}{\pi} \right)^{n/2} \frac{1}{(A')^{n/2-1}} \int_0^\infty J_{n/2-1}(A'r) \frac{r^{n/2+1}}{\sinh(r)} dr \quad (53)$$

where J_ν is a Bessel function. Closed form expressions can be given for odd n values. For instance:

$$n = 3 \quad P(A') = \frac{3\pi}{2A'} \frac{\tanh(\pi\sqrt{3}A')}{\cosh^2(\pi\sqrt{3}A')} \quad (54)$$

$$n = 5 \quad P(A') = \frac{5}{4A'^3} \frac{\tanh(\pi\sqrt{5}A') - (\pi\sqrt{5}A')(1 - 3 \tanh^2(\pi\sqrt{5}A'))}{\cosh^2(\pi\sqrt{5}A')} \quad (55)$$

Now, if we consider the distribution of the sum of the areas $\mathcal{A} = \sum_{i=1}^n A_i$, it is obtained by setting $B_j = B, \forall j$. (52) leads to ($\mathcal{A}' = \mathcal{A}/t$):

$$\langle e^{iB\mathcal{A}'} \rangle = \frac{B}{2 \sinh\left(\frac{B}{2}\right)} \quad (56)$$

With (5), we see that the sum of areas has, asymptotically, exactly the same distribution as the area enclosed by a single Brownian particle. In fact, we can compute $\langle e^{iB\mathcal{A}'} \rangle$ for all t values (and not only when $t \rightarrow \infty$). This is because, that time, the matrix \mathbf{B} ($\mathbf{B}_{ij} = B \delta_{ij}$) commutes with \mathbf{M} . So, we are left with an { harmonic oscillator + uniform magnetic field } problem for each normal coordinate (except for Z_1 , that only feels a pure magnetic field). We get the result [14]:

$$\langle e^{iB\mathcal{A}'} \rangle = \frac{B}{2 \sinh\left(\frac{B}{2}\right)} \prod_{i=2}^n \frac{F_i(B)}{F_i(0)} \quad (57)$$

$$F_i(B) = \frac{\omega'_i}{2\pi \sinh(t\omega'_i)} \exp\left(-\frac{\omega'_i}{2\pi \sinh(t\omega'_i)} (\cosh(t\omega'_i) - \cosh(B/2)) |Z_i^{(0)}|^2\right) \quad (58)$$

$$\omega'_i = \sqrt{\omega_i^2 + \left(\frac{B}{2t}\right)^2} \quad (59)$$

We recover (56) in the limit $t \rightarrow \infty$ ($\prod_{i=2}^n F_i(B)/F_i(0) \rightarrow 1$ when $t \rightarrow \infty$).

To close this section, it is interesting to consider the asymptotic law for the area A'_j ($= A_j/t$) enclosed by a given monomer j . (52) gives:

$$\langle e^{iB_j A'_j} \rangle = \frac{B_j}{2\sqrt{n} \sinh\left(\frac{B_j}{2\sqrt{n}}\right)} \quad (60)$$

It follows that A_j satisfies Lévy's law (5) and scales like $\frac{t}{\sqrt{n}}$. Remark that the area swept by the chain c.o.m., G , should scale like $\frac{t}{n}$. On the other hand, for the same gaussian noise, we would get $A_j \sim t$ if particle j was free (i.e. $k = 0$). The actual scaling of A_j is intermediate: this particle moves more freely than G but it is embedded in the chain, thus not completely free!

Those considerations allow us to give a more precise sense to the statement: “the process is dominated by the c.o.m. motion”. This one is true as long as we look at occupation times. However, when we study finer quantities like areas, this sentence must be corrected. Similar (even more dramatic) deviations will occur when we look at winding angles.

To end up with areas, let us remark that the case of open trajectories can be treated exactly along the same lines as the one developed here, without additionnal difficulties (in particular, (48) still holds). We will not address this problem in the present work.

5 Winding angles distribution

The last part of this paper will be devoted to the distribution $P(\{\theta_j\})$ (θ_j is the angle wound around O by particle j during a time t). We consider the same conditions as for Spitzer's law ($z^{(0)}$, initial configuration, fixed, with $z_j^{(0)} \neq 0, \forall j$; z , final configuration, unspecified; $t \rightarrow \infty$).

We want to proceed as before and insert the constraint:

$$\prod_{j=1}^n \delta \left(\theta_j - \frac{1}{2i} \int_0^t \left(\frac{z_j \ddot{z}_j - \bar{z}_j \dot{z}_j}{z_j \bar{z}_j} \right) d\tau \right) \quad (61)$$

in the Wiener measure (15). We are now faced with the problem of n harmonically bound particles submitted to the magnetic fields of n different point-like vortices located at the origin. The corresponding hamiltonian is:

$$H_\lambda = H_0 + V_\lambda \quad (62)$$

$$V_\lambda = \sum_{i=1}^n \lambda_i \left(\frac{1}{z_i} \partial_{\bar{z}_i} - \frac{1}{\bar{z}_i} \partial_{z_i} \right) + \sum_{i=1}^n \frac{\lambda_i^2}{2z_i \bar{z}_i} \quad (63)$$

and the distribution $P(\{\theta_i\})$ is given by:

$$P(\{\theta_i\}) = \int \left(\prod_{j=1}^n \frac{d\lambda_j}{2\pi} e^{i\lambda_j \theta_j} \right) \int dz d\bar{z} F(z, z^{(0)}) G_\lambda(z, z^{(0)}, t) \quad (64)$$

$$G_\lambda(z, z^{(0)}, t) = \left\langle z \left| e^{-tH_\lambda} \right| z^{(0)} \right\rangle \quad (65)$$

Studying the limit $t \rightarrow \infty$, we cannot develop directly as before a perturbation theory with V_λ : this is because the last term in V_λ gives a divergent contribution [9]. Due to this term, all the eigenfunctions of H_λ must vanish in O at least as $\prod_{i=1}^n |z_i|^{|\lambda_i|}$ ($\equiv U(z)$). So, we redefine those eigenfunctions [9]:

$$\Psi = U \tilde{\Psi} \quad (66)$$

The new hamiltonian acting on $\tilde{\Psi}$ is

$$\widetilde{H}_\lambda = H_0 + \widetilde{V}_\lambda \quad (67)$$

$$\widetilde{V}_\lambda(z) = \sum_{i=1}^n \left((\lambda_i - |\lambda_i|) \frac{1}{z_i} \partial_{\bar{z}_i} - (\lambda_i + |\lambda_i|) \frac{1}{\bar{z}_i} \partial_{z_i} \right) \quad (68)$$

with a propagator \widetilde{G}_λ

$$\widetilde{G}_\lambda(z, z^{(0)}, t) = \left\langle z \left| e^{-t\widetilde{H}_\lambda} \right| z^{(0)} \right\rangle = \frac{U(z^{(0)})}{U(z)} G_\lambda(z, z^{(0)}, t) \quad (69)$$

($\widetilde{G}_0 = G_0$). That time, the perturbation theory is properly defined and we can compute the characteristic function:

$$C(\{\lambda_j\}) \equiv \left\langle e^{i \sum_{j=1}^n \lambda_j \theta_j} \right\rangle = \int dz d\bar{z} \left(\prod_{j=1}^n \frac{|z_j|^{| \lambda_j |}}{|z_j^{(0)}|^{| \lambda_j |}} \right) F(z, z^{(0)}) \widetilde{G}_\lambda(z, z^{(0)}, t) \quad (70)$$

with, symbolically,

$$\widetilde{G}_\lambda = \sum_{m=0}^{\infty} (-1)^m G_0^\infty (\widetilde{V}_\lambda G_0^\infty)^m \quad (71)$$

Using integration by parts and also the relationship $\partial_{z_i} \left(\frac{1}{\bar{z}_i} \right) = \pi \delta(z_i)$, we first calculated $C(\{\lambda_i\})$ up to 4th order in \widetilde{V}_λ , with the result:

$$C(\{\lambda_j\}) \sim e^{X/2} D(X) \quad (72)$$

$$D(X) = 1 + \left(\frac{n+1}{2} \right) \left(\frac{-X}{1!} + \frac{X^2}{2!} n - \frac{X^3}{3!} \left(\frac{3n^2-1}{2} \right) + \frac{X^4}{4!} (3n^3 - 2n) - \dots \right) \quad (73)$$

$$X = \left(\sum_{i=1}^n |\lambda_i| \right) \ln t \quad (74)$$

The prefactor $e^{X/2}$ comes out from $U(z)$ in (70) when integrated over the final configuration: it will be present at all orders of the computation. Moreover, (72) suggests that $C(\{\lambda_i\})$ is only a function of X : this is actually the case, as will be shown in the sequel.

Let us consider the m^{th} order term in (70,71) and suppose that we integrate, first, over $z, z^{(m)}, z^{(m-1)}, \dots, z^{(k+1)}$. Following the computation step by step, it is not difficult to convince oneself that the integration over $z^{(k)}$ involves expressions of the form:

$$\int d\bar{z}^{(k)} dz^{(k)} \phi(z^{(k)}, T) \widetilde{V}_\lambda(z^{(k)}) G_0^\infty(z^{(k)}, z^{(k-1)}, t_k - t_{k-1}) \quad (75)$$

$$\text{where } \phi(z^{(k)}, T) = e^{-\frac{|Z_1^{(k)}|^2}{2T}} e^{-\frac{1}{2} \sum_{i=2}^n k \omega_i |Z_i^{(k)}|^2} \quad (76)$$

and $T = t_l - t_k, k+1 \leq l \leq m$. Let us call J_k the result of (75). In the limit of long times, it reads:

$$J_k = - \left(\sum_{i=1}^n |\lambda_i| \right) \times \left(-\frac{n+1}{2(t_k - t_{k-1})} \phi(z^{(k-1)}, t_k - t_{k-1}) + \frac{1}{T + t_k - t_{k-1}} \phi(z^{(k-1)}, T + t_k - t_{k-1}) \right) \quad (77)$$

The m successive spatial integrations produce the factor $(\sum |\lambda_i|)^m$ and, at the end, we are left with time integrals of the form:

$$I_m(i_{m-1}, \dots, i_0)(t) = \int_0^t dt_m \int_0^{t_m} dt_{m-1} \dots \int_0^{t_2} dt_1 \frac{e^{-\sum_{i=1}^m \frac{\alpha_i}{t_i} - \sum_{i=1}^{m-1} \frac{\beta_i}{t_{i+1} - t_i}}}{(t_{i_{m-1}} - t_{m-1}) \dots (t_{i_1} - t_1) t_{i_0}} \quad (78)$$

with $\alpha_i, \beta_i > 0$ and

$$i_{m-1} = i_{m-2} = \dots = i_k = m ; i_{k-1} = i_{k-2} = \dots = i_l = k ; i_{l-1} = i_{l-2} = \dots = i_j = l ; \dots \blacksquare \quad (79)$$

We have proved, step by step, that;

$$I_m(i_{m-1}, \dots, i_0)(t) \sim_{t \rightarrow \infty} \frac{(\ln t)^m}{\prod_{l=0}^{m-1} (i_l - l)} \quad (80)$$

Those considerations show that, actually, $C(\{\lambda_i\})$ is only a function of X . So, we can write $D(X) = \sum_{m=0}^{\infty} a_m X^m$, with $a_0 = 1$ (see eq.(73)).

Moreover, with the help of the above equation (80), and also looking at the tree structure exhibited in eq.(77), the following recursion relation can be shown:

$$a_m = y \sum_{k=0}^{m-1} \frac{a_k}{(m-k)!} \quad (81)$$

$$y = -\frac{n+1}{2} \quad (82)$$

It allows to write a closed form formula for $D(X)$:

$$D(X) = \frac{1}{1 - y(e^X - 1)} = \frac{e^{-X/2}}{\cosh(X/2) + n \sinh(X/2)} \quad (83)$$

With (72) and, also, a rescaling of the angles ($\theta'_i = \frac{2\theta_i}{\ln t}$), we get the desired characteristic function:

$$\left\langle e^{i \sum_{j=1}^n \lambda_j \theta'_j} \right\rangle = \frac{1}{\cosh(u) + n \sinh(u)} \quad (84)$$

$$u = \sum_{i=1}^n |\lambda_i| \quad (85)$$

(with $n = 1$, we recover eq.(3)).

We consider (84) as the main result of this paper.

Finally, Fourier transformation shows that $P(\{\theta'_j\})$ is an “infinite sum of products of Spitzer’s laws” (!) with highly correlated variables:

$$P(\{\theta'_j\}) = \frac{2}{n+1} \sum_{k=0}^{\infty} \left\{ \left(\frac{n-1}{n+1} \right)^k \left(\prod_{j=1}^n \frac{1}{\pi(2k+1)} \frac{1}{1 + \left(\frac{\theta'_j}{2k+1} \right)^2} \right) \right\} \quad (86)$$

All the moments of this distribution are infinite (unless they trivially vanish).

For a given particle j of the chain, we have:

$$\langle e^{i\lambda_j\theta'_j} \rangle = \frac{1}{\cosh(\lambda_j) + n \sinh(|\lambda_j|)} \quad (87)$$

that leads to:

$$P(\theta'_j) = \frac{2}{n+1} \sum_{k=0}^{\infty} \left\{ \left(\frac{n-1}{n+1} \right)^k \frac{1}{\pi(2k+1)} \frac{1}{1 + \left(\frac{\theta'_j}{2k+1} \right)^2} \right\} \quad (88)$$

The difference with Spitzer's law is due to the presence of n in the denominator of (87).

To shed some light on this problem, let us go back to the joint law (6) of small and big windings for the chain c.o.m.. What could we expect for the corresponding windings of particle j ? With little effort, we can say that:

- i) The big windings will be roughly the same for both (when the chain is far from O, particle j follows the c.o.m. and winds around O in the same way). So, we keep λ_+ unchanged in (6).
- ii) The small windings will be quite different. This is because particle j is artificially maintained in the vicinity of O: despite its higher mobility, *it spends the same time* as the c.o.m. in a given domain surrounding O. As a consequence, its small windings law will be broadened. Assuming that the remark following (7) holds, we get this broadening by changing $|\lambda_-|$ into $n|\lambda_-|$ in (6) (n is the ratio of the diffusion constants; of course, we don't say at all that (7) is the law of small windings!).

Thus, our guess for particle j is:

$$\langle e^{i(\lambda_+\theta'_{j+} + \lambda_-\theta'_{j-})} \rangle = \frac{1}{\cosh(\lambda_+) + n \frac{|\lambda_-|}{\lambda_+} \sinh(\lambda_+)} \quad (89)$$

Setting $\lambda_+ = \lambda_- = \lambda_j$, we recover (87). We are aware that this argument is strictly heuristic and that (89) remains to be proved. Nevertheless, we think that it allows to explain correctly the presence of n in (87).

6 Conclusion

Let us briefly summarize this work. We have computed explicitly the asymptotic joint laws of the occupation times, areas and winding angles of a chain of harmonically bound Brownian particles.

For all these properties, we have shown that the scaling variables take the same *general* form as for the standard Brownian motion. However, a detailed study reveals

important specific features that reflect a subtle interplay between the free c.o.m. motion – that strongly influences the whole chain properties – and the relative freedom of a given particle of the chain. For occupation times distributions, it appears that the c.o.m. satisfies the same law as a given monomer; now, for the areas, the scaling becomes slightly different and, finally, for the winding angles, the law itself is changed. Remark also that correlations are systematically present.

Moreover, we observe that the scaling variables and the laws are very different from those met in our study of the attached Rouse chain [15] (in the latter case, $\theta \sim t$, $A \sim \sqrt{t}$, and the winding angles are uncorrelated). These differences are not so surprising since, in that case, we had no translation invariance.

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References

- [1] Rouse P E 1953 *J. Chem. Phys.* **21** 1273
- [2] de Gennes P G 1979 *Scaling Concepts in Polymer Physics* (Ithaca)
- [3] Doi M and Edwards S F 1986 *The Theory of Polymer Dynamics* (Oxford)
Grosberg A Y and Khokhlov A R 1994 *Statistical Physics of Macromolecules* (New-York, translated from russian)
- [4] Kallianpur G and Robbins H 1953 *Proc. Nat. Acad. Sci. U.S.A.* **39** 525
- [5] Spitzer F 1958 *Trans. Am. Math. Soc.* **87** 187
- [6] Lévy P 1948 *Processus Stochastiques et Mouvement Brownien* (Paris)
- [7] Pitman J W and Yor M 1986 *Ann. Prob.* **14** 733; 1989 **17** 965
- [8] Messulam P and Yor M 1982 *J. Lond. Math. Soc. (2)* **26** 348
Rudnick J and Hu Y 1987 *J. Phys. A: Math. Gen.* **20** 4421
Berger M A 1987 *J. Phys. A: Math. Gen.* **20** 5949
Brereton M G and Butler C 1987 *J. Phys. A: Math. Gen.* **20** 3955
- [9] Comtet A, Desbois J and Monthus C 1993 *J. Phys. A: Math. Gen.* **26** 5637
- [10] van Kampen N G 1981 *Stochastic Processes in Physics and Chemistry* (Amsterdam)
- [11] Riesz F and Sz-Nagy B 1955 *Functional Analysis* (Frederick Ungar Publ. Co.)
- [12] Feynman R P and Hibbs A R 1965 *Quantum Mechanics and Path Integrals* (New-York)
- [13] Hughes B D 1995 *Random Walks and Random Environments* (Oxford)
- [14] de Veigy A and Ouvry S 1992 *Nucl. Phys. B* **388** 715
- [15] Bénichou O and Desbois J 2000 submitted to *J. Stat. Phys.*

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